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MOLECULAR-DYNAMICS INVESTIGATION OF NONLINEAR DIELECTRIC RESPONSE FOR A FERROELECTRIC MODEL

By

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Abstract

The relation between the applied electric field E and it's response P of a unified classical oscillator model for order-disorder and displacive ferroelectrics is investigated numerically on computer. It is shown that the relation between the E and the imaginary part of P depends on the type of the ferroelectrics. On the other hand, there are few differences between the two types in the static field case of the E-P relation.

Recently, Onodera[1] has calculated the dynamic susceptibility of a unified oscillator model for oder-disorder and displacive ferroelectrics with the aid of the linear-response theory.[2),3)] He considered an assembly of classical oscillators moving in the anharmonic potential $V(x)=Ax^4+Bx^2$, where x stands for the displacement of an oscillator. A is taken to be definitely positive, while B may be either positive or negative. The potential has one or two minima, depending on the sign of B. An interaction between these anharmonic oscillators is bilinear in their displacements and the interaction is treated in the Weiss approximation. The applied electric field E interacts with the dipole moment of the oscillator, which he assumes to be proportional to it's displacement.

In the present letter, we shall numerically investigate the nonlinear dielectric response of a ferroelectric model which is similar to Onodera's model. Our model can be described by the Hamiltonian.

$$H=\sum_{i=1}^{N}\left\{\frac{1}{2}M\dot{x}_i^2+(Ax_i^4+Bx_i^2)+\gamma\left(\frac{1}{N-1}\sum_{j\neq i}^{N}x_j\right)x_i + E\cos(\omega t)\,x_i\right\}+H', \tag{1}$$

Where N is the number of the oscillators, M is the mass of the oscillator, γ is the coupling constant and ω is the angular frequency of the applied electric field. H' is the coupling Hamiltonian between the system and a heat bath which is expressed by an ideal gas. We take the mass of the gas's particle 0.1 times as heavy as the mass of the

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oscillator and the particle of the gas collides with the oscillator. The collision produces a friction of the motion of the oscillator. The friction is a loss mechanism of our model which is not contained in Onodera's model. If we take the thermodynamic limit, namely N tend to infinity, our model undergoes a second-order phase transition in both $B>0$ and $B<0$ cases at a Curie temperature determined by γ[1].

We define the electric polarization P as $P=(\sum_{i=1}^{N} x_i)/N$. The time dependence of P can be expressed as

$$P(t) = P' \cos(\omega t) + P'' \sin(\omega t) , \qquad (2)$$

where we neglect the higher harmonic terms.

According to Onodera we use the following units:

$$\text{Temperature} \cdots\cdots B^2/4Ak_B ,$$

$$\text{Frequency} \cdots\cdots (2|B|/M)^{1/2} ,$$

where k_B is the Boltzmann constant.

We investigate the time evolution of the system by solving the equations of motion of the Hamiltonian $(H-H')$ numerically on computer by means of the Runge-Kutta method. The collision processes are inserted in the above time evolution about 10 times in a period of the oscillation in which we use random numbers. The random number expresses the velocity of the particle v of the one-dimensional ideal gas. Making use of the equipartition law of energy and $\bar{v}=0$, the standard deviation of the velocity is determined by the temperature of the heat bath T and the mass of the particle m as $\{\overline{(v-\bar{v})^2}\}^{1/2}=\{\overline{v^2}\}^{1/2}=\{k_BT/m\}^{1/2}$. We calculate P' and P'' as a long time average, therefore, our results do not depend on the initial condition of our model. Our computer simulations show that the dependence of N is so small that we take $N=40$.

The polarization P versus the applied electric field E in displacive case, namely $B>0$, at several temperatures with $\omega=0$ is shown in Fig. 1. The same relation of the order-disorder case, namely $B<0$, is shown in Fig. 2. Fig. 1 and 2 show that there are few differences between the order-disorder case and the displacive case in the static nonlinear dielectric response. The E-P relation with $\omega=0.25$ of the displacive case and order-disorder case are shown in Fig. 3 and 4, respectively. In both cases, the temperatures are taken near their Curie points which are estimated by Fig. 1 and 2. It is interesting that in dynamic case, the relation E-P'' depends on the type of the ferroelectrics.

Onodera's model becomes non-ergodic when $B<0$.[1] It is a difficult question whether we can apply the linear-response theory to the non-ergodic case or not. In our case, however the coupling Hamiltonian H' enables us to remove the problem of the non-ergodicity. In usual cases, the motion of the oscillation accompanies a friction which gives a loss. Due to the convenience of the calculation, we express the friction by H'.

Further studies including the friction dependence of the response are now gogin

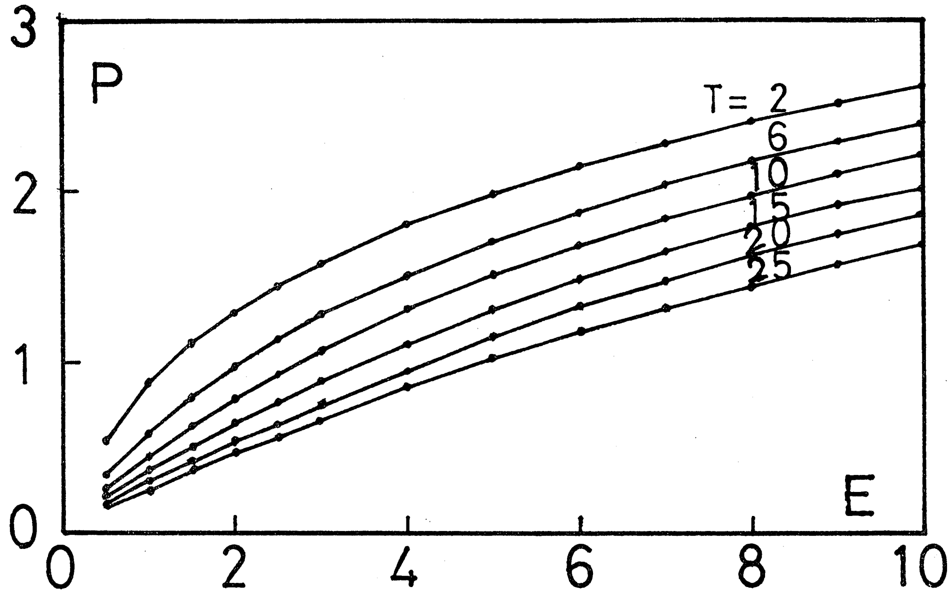

Fig. 1. The static E-P relation of the displacive case with A=1, B=2, γ=−2 and N=40 at several temperatures.

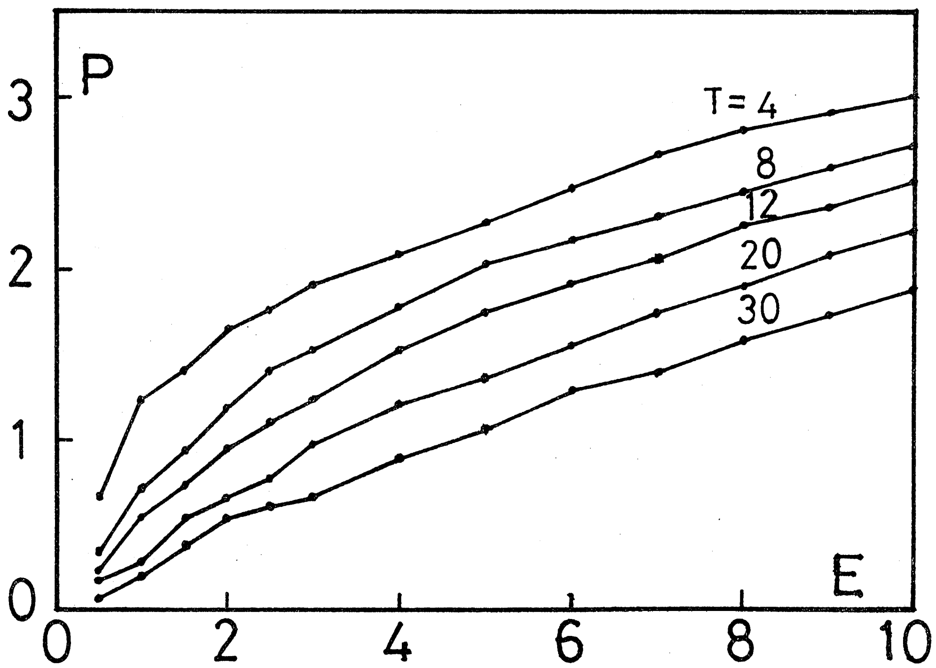

Fig. 2. The static E-P relation of the order-disorder case with A=1, B=−2, γ=−0.1 and N=40 at several temperatures.

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The numerical calculations were performed using a FACOM 230–75 computer at the Kyushu University Computing Center and a FACOM 230–45S computer at the Kagoshima University Computing Center.

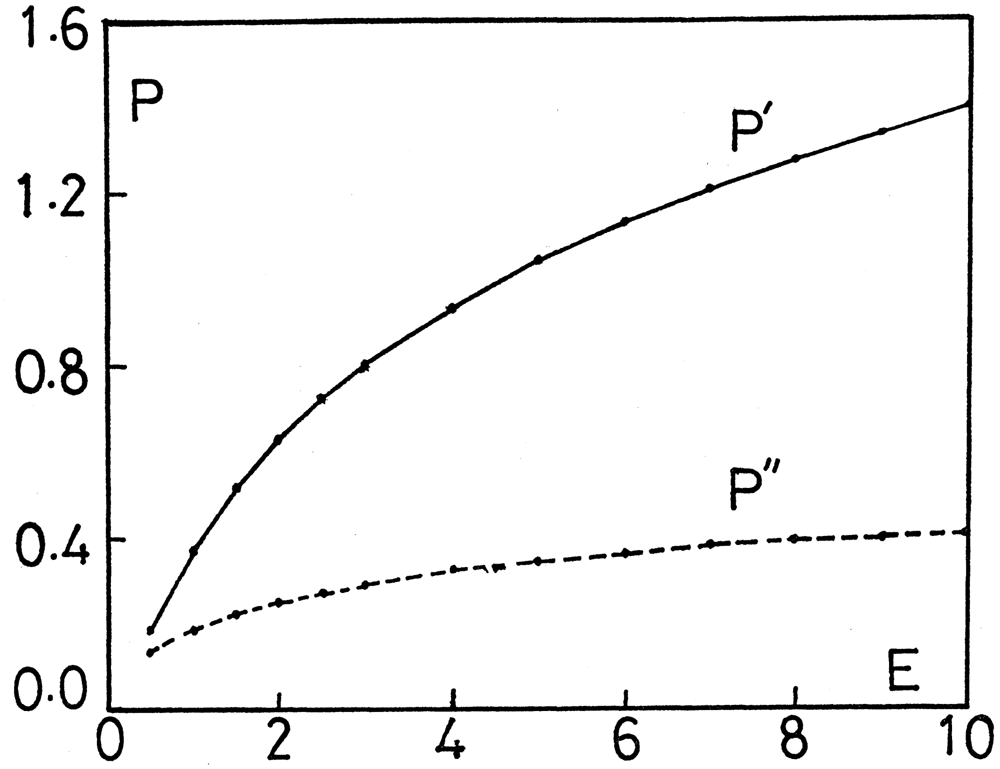

Fig. 3. The E-P' and E-P'' relations of the displacive case with $A=1$, $B=2$, $\gamma=-2$, $N=40$ and $\omega=0.25$ at $T=2$.

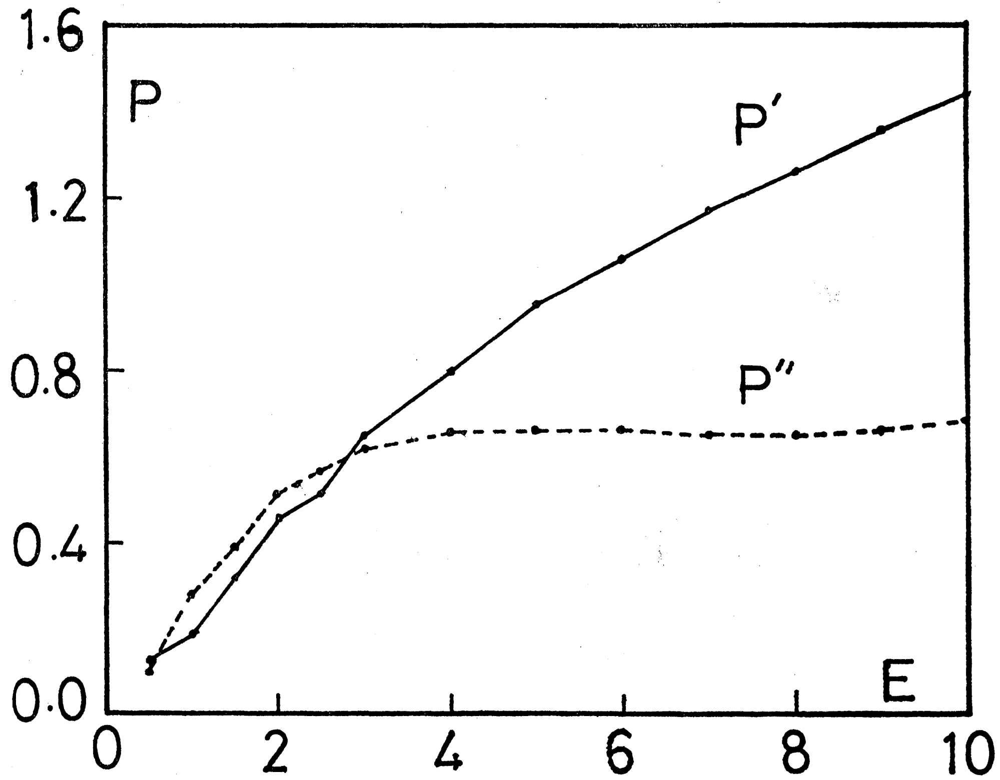

Fig. 4. The E-P' and E-P'' relations of the order-disorder case with $A=1$, $B=-2$, $\gamma=-0.1$, $N=40$ and $\omega=0.25$ at $T=4$.

References

1) Y. Onodera: Progr. theor. Phys. **44** (1970) 1477.
2) R. Kubo: J. Phys. Soc. Japan **12** (1957) 570.
3) D.N. Zubarev: Uspekhi Fiz. Nauk SSSR **71** (1960) 71.